(51) International Patent Classification 7 :		(11) International Publication Number:	**WO 00/30202**
H01M 8/10	**A1**	(43) International Publication Date:	25 May 2000 (25.05.00)

(72) Inventors: BRAUN, James, C.; 1975 Wheeler Road, Juno Beach, FL 33408 (US). ZABRISKIE, John, E., Jr.; 1841 SW Effland Avenue, Port St. Lucie, FL 34953 (US). NEUTZLER, Jay, K.; 3204 Morning Glory Court #104, Palm Beach Gardens, FL 33410 (US). FUCHS, Michel; 9 Via de Casas Norte, Boynton Beach, FL 33426 (US). GUSTAFSON, Robert, C.; 4297 Express Street, Palm Beach Gardens, FL 33410 (US).

(74) Agents: NELSON, Gregory, A. et al.; Quarles & Brady LLP, 4th floor, 222 Lakeview Avenue, P.O. Box 3188, West Palm Beach, FL 33402–3188 (US).

(54) Title: FUEL CELL COLLECTOR PLATE AND METHOD OF FABRICATION

(57) Abstract

An improved molding composition is provided for compression molding or injection molding a current collector plate for a polymer electrolyte membrane fuel cell. The molding composition is comprised of a polymer resin combined with a low surface area, highly–conductive carbon and/or graphite powder filler. The low viscosity of the thermoplastic resin combined with the reduced filler particle surface area provide a moldable composition which can be fabricated into a current collector plate having improved current collecting capacity vis–a–vis comparable fluoropolymer molding compositions.

FUEL CELL COLLECTOR PLATE AND METHOD OF FABRICATION

Field of the Invention

This invention relates generally to compositions and methods for

5   fabricating electrically-conductive polymer composite structures and

coatings, and more particularly to a highly-conductive graphite composite

particularly suited for net shape molding a current collector plate for a fuel

cell.

10   Background of the Invention

Solid polymer electrolyte membrane (PEM) type electrochemical fuel

cells are well known. Generally, PEM fuel cells comprise a membrane

electrode assembly (MEA) and diffusion backing structure interposed

between electrically conductive graphite current collector plates. In

15   operation, multiple individual cells are arranged to form a fuel cell stack.

When the individual cells are arranged in series to form a fuel cell stack, the

current collector plates are referred to as bipolar collector plates. The

collector plates perform multiple functions, including: (1) providing structural

support; (2) providing electrical connection between cells; (3) directing fuel

20   and oxidant reactants and/or coolant to individual cells; (4) distributing

reactant streams and/or coolant within individual cells; (5) removing

byproduct from individual cells; and (6) separating fuel and oxidant gas

streams between electrically connected cells. In addition to being electrically

conductive, collector plates must have good mechanical strength, high

25   thermal stability, high resistance to degradation caused by chemical attack

and/or hydrolysis, and low permeability to hydrogen gas.

Typically, collector plates have intricate patterns formed on their major

surfaces. For instance, integral channels may be provided for directing fuel,

oxidant and/or byproduct through the fuel cell. Historically, graphite

30   structures have been machined to a desired configuration from graphite

composite blanks. Due in part to the expense and time consuming nature of

machining, more recent efforts in the fuel cell manufacturing industry have focused on the development of compositions and methods for producing net shape molded fuel cell structures, such as bipolar collector plates, using compression molding and injection molding techniques. These efforts, which

5   have had limited success, have concentrated primarily on molding compositions incorporating fluoropolymer binder materials. For example, bipolar collector plates molded from thermoplastic fluoropolymers, such as vinylidene fluoride, are disclosed in U.S. Patent Nos. 3,801,374, 4,214,969, and 4,988,583.

10   Compared to other polymeric materials, fluoropolymers have relatively high viscosities. Significantly, the relatively high viscosity associated with fluoropolymers limits their effectiveness as binder materials in molding and coating compositions.

In an effort to maximize the electrical conductivity of current collector

15   plates for fuel cells, it is desirable to maximize electrically-conductive filler loading levels. Generally, as the percentage of filler particles in a given polymer composition is increased, there is a corresponding increase in the viscosity of the composition. Consequently, regardless of the polymer binder material chosen, the addition of electrically conductive filler must be

20   limited to ensure some minimum degree of flow during processing. Such viscosity limitations are particularly pronounced in injection molding applications, where the viscosity of the polymer composition must be maintained at a low enough level to allow the composition to travel through intricate mold features such as channels and gates. In the case of

25   fluoropolymer compositions, the high initial viscosity level associated with the fluoropolymer binder restricts the quantity of filler that can be loaded into the binder prior to processing. Consequently, the electrical conductivity of fuel cell collector plates fabricated using fluoropolymer binders is correspondingly limited.

30   For these and other reasons, there is a well-established need for improved compositions and methods for processing highly conductive

composite structures for electronic, thermoelectric and electrochemical
device applications.

Summary of the Invention

It is an object of this invention to provide a composition for fabricating thermally- and electrically-conductive polymer composite structures and

5   coatings for use in highly-corrosive environments, wherein the electrical conductivity of the resulting structure or coating is improved as a result of enhanced filler loading capacity of the composition.

It is another object of this invention to provide a composition, and a method for processing said composition, to form a thermally- and

10   electrically-conductive polymer composite structure or coating for use in electronic, thermoelectric and electrochemical devices.

It is another object of this invention to provide a non-fluorinated composition for rapidly net shape molding a current collector plate for a polymer electrolyte membrane (PEM) fuel cell, wherein improved filler loading

15   results in a current collector plate having a higher bulk electrical conductivity than conventional current collector plates fabricated from fluoropolymer-based compositions.

These and other objects of the invention are achieved with the novel compositions and methods of the present invention. Novel polymer

20   compositions are provided for producing highly-conductive coatings and net shape molded structures for a variety of applications, including: corrosion-resistant electrical and thermal conductors and contacts; battery and capacitor electrodes; electrodes for electrochemical coating and synthesis of materials; and electrochemical device components, such as current collector

25   plates for polymer electrolyte membrane (PEM) fuel cells.

Briefly, according to the invention, a highly-loaded polymer composition is provided for fabricating a structure or coating generally suitable for use in electronic, thermoelectric and electrochemical devices. In the preferred embodiment of the invention, the composition is particularly

30   suited for compression molding and/or injection molding a current collector plate for a PEM fuel cell. The composition is comprised of a low viscosity

4

polymer loaded with a chemically-inert, thermally and electrically conductive filler.

The polymer is chosen from the group of polymers having a melt viscosity of less than 1,000 Newton-seconds per square meter ($N*s/m^2$) over a shear rate range of 1,000 to 10,000 sec^{-1}. Furthermore, it is preferred that the polymer has material properties and characteristics as summarized in Table 2 (below). Suitable families of polymers include: polyphenylene sulfide (PPS); modified polyphenylene oxide (PPO); liquid crystal polymer (LCP); polyamide; polyimide; polyester; phenolic; epoxy-containing resin and vinyl ester.

The polymer composition is loaded with highly-conductive filler. In the preferred embodiment of the invention, the filler comprises carbon and/or graphite particles having an average particle size ranging from approximately 0.1 to 200 microns, and preferably in the range of about 23 to 26 microns. The filler particles have a surface area ranging from approximately 1 to 100 m^2/g, and preferably in the range of 7 to 10 m^2/g (as measured by BET testing standards). The composition may include additional components, including: carbon and/or graphite nanofibers; carbon and/or graphite fibers; metal fibers such as stainless steel or nickel; and metal-coated carbon and/or graphite fiber concentrates having thermoplastic or thermoset sizing chosen from the aforementioned group of potential polymers.

The composition is subsequently formed into a desired shape by compression molding, injection molding, or a combination thereof. Alternatively, the composition can be used in cladding or coating operations.

Detailed Description of the Preferred Embodiments

A novel composition is provided for fabricating a corrosion-resistant composite or surface coating having improved electrical conductivity. In the best mode of the invention, the composition is used to mold a unitary current collector plate for a polymer electrolyte membrane (PEM) fuel cell. However, the composition can be used to fabricate alternative collector plate structures as well. For instance, the composition can be coated onto the surface of a suitable substrate to form a multilayer collector plate structure. Accordingly, the term "structure" as used herein is intended to refer to either a unitary part or a coated part. Preferably, the composition comprises a low viscosity thermoplastic resin combined with a highly-conductive carbon or graphite filler material.

The composition is chosen to produce a current collector plate capable of withstanding the harsh environment of a PEM fuel cell. Preferably, the composition is used to fabricate a current collector plate meeting particular criteria listed in Table 1 (below). In addition to having the properties and characteristics identified below, it is preferred that the collector plate is resistant to chemical and electrochemical degradation and hydrolysis, and has a bulk electrical resistance less than 50 mΩ-cm (or a bulk conductivity greater than 20 S/cm).

Property	Test Method	Value	Comments
Bulk Resistance	4-point probe	<50 mΩ-cm	
Bulk Density		1.5-2.25 g/cc	>2.25 (coated metals)
H_2 Permeability		<5$(10)^{-6}$ cm/s	90°C; 202(10^3)N/m^2
Thermal Index	UL746B	>45°C	tensile strength

Table 1

Suitable binder resins are defined as non-fluorinated thermoplastic or thermoset polymers preferably having melt viscosities of less than 1,000 Newton-seconds per square meter ($N*s/m^2$) over a shear rate range of 1,000 to 10,000 sec^{-1}, and additional material properties and characteristics

5   defined in Table 2 (below). As used herein, the term "non-fluorinated" is intended to describe polymers other than fluoropolymers. Accordingly, nominal quantities of fluorine-containing components may be added to the present composition without changing the designation of the binder resin as a non-fluorinated polymer. For example, nominal quantities of Teflon® may

10   be added to the binder resin to improve mold release characteristics of the final composition.

Property	Method	Value	Comments
Viscosity	Capillary Rheometry	< 1,000 $N*s/m^2$	over a shear rate of 1,000-10,000 sec^{-1}
Thermal Index	UL746B	>45°C	tensile strength
Hydrolytic Stability		>80% property retention	60°C water; 1,000 hr
Particle Size	60 mesh screen	>50% by wt.	<200 microns
$T_{HEAT\ DEFLECTION}$	ASTM D648	>75°C	at $1.82(10)^6\ N/m^2$
$T_{MELTING}$		>90°C	pref. 315-340°C
Tensile Strength	ASTM D638	$21-210(10)^6\ N/m^2$	pref. $>40(10)^6\ N/m^2$
Density		1.0-2.0 g/cc	
Water Absorption	ASTM D570	<10% wt gain	23°C; 24 hours

Table 2

Particular examples of polymer resins which meet these requirements include, but are not limited to, polyphenylene sulfide (PPS), low molecular

30   weight PPS, liquid crystal polymer (LCP), and modified polyphenylene oxide.

Suitable polyphenylene sulfides are commercially available from Phillips

Chemical Company of Bartlesville, Oklahoma, under the trade name Ryton®,

and from Ticona Corporation of Summit, New Jersey, under the trade name

Fortron®. Liquid crystal polymers having the desired properties are

5   commercially available from Ticona under the trade name Vectra®, and from

Amoco Performance Products, Inc. of Alpharetta, Georgia, under the trade

name Xydar®. A modified polyphenylene oxide having the desired properties

is commercially available from General Electric Company of Pittsfield,

Massachusetts, under the trade name Noryl®. Combinations of the above-

10   identified polymer resins have the desired properties listed in Table 2.

Prior to being molded, the polymer resin is combined with highly

conductive filler particles. Preferably, the filler particles comprise carbon

and/or graphite and have properties and characteristics as defined below in

Table 3.

15

Property	Method	Value	Comments
Carbon Content	--	>89%	ideal: >98%
Pressed Density	--	1.8-2.0 g/cm^3	at 44.8(10)^6 N/m^2
Particle Size	200 mesh screen	>70% by wt.	ideal: >98% by wt.
Average Particle Size	--	0.1-200 μm	ideal: 23-26 μm
Surface Area	BET	5-50 m^2/g	Ideal: 7-10 m^2/g
Electrical Resistivity	--	<15 mΩ-cm	at 48(10)^6 N/m^2

25

Table 3

The filler may be provided in various forms, including powder, fiber

and flake. However, it is preferred that the filler material comprises a high

8

purity graphite powder having a carbon content of greater than 98 percent. The use of graphite is preferred because graphite is electrochemically stable in a wide range of environments. The use of a powder form is preferred because powders are less apt to impede the flow of the composition during

5   molding. Preferably, the graphite powder has an average particle size of approximately 23-26 microns, and a BET-measured surface area of approximately 7-10 m^2/g. The incorporation of small, low surface area conductive particles in the novel composition of the present invention is a significant departure from conventional conductive composites used to

10   fabricate structures for electronic, thermoelectric and electrochemical devices. Conventional conductive composites, such as those used to fabricate fuel cell collector plates, typically contain conductive particles having a very high surface area combined with a small particle size. For instance, carbon black particles having a surface area of greater than 500

15   m^2/g and a particle size of less than 1 micron are typical. Commonly, conventional conductive composites also contain large fibers having a low surface area. For instance, fibers having a surface area of less than 10 m^2/g coupled with a fiber length in excess of 250 microns are typical.

The combination of reduced filler particle size and reduced filler

20   particle surface area provides a means for maintaining material flow while increasing filler particle loading. Significantly, the relatively low particle size and surface area enable greatly improved filler particle packing densities as compared to known compositions for molding current collector plates. A corresponding increase in solids loading results in a fabricated plate having

25   increased electrical conductivity, while minimizing gas permeable voids. Graphite powders having the above-identified properties are available from UCAR Carbon Company, Inc. of Lawrenceburg, Tennessee, as well as from Asbury Carbons, Inc. of Asbury, New Jersey.

Carbon nanofibers may be added to the composition to improve

30   electrical conductivity and mechanical strength of the molded collector plate. The carbon nanofibers typically have diameters ranging from a few

nanometers to several hundred nanometers, and aspect ratios ranging from
50 to 1,500. Further additives may include carbon fibers, metal fibers such
as stainless steel or nickel, and/or metal-coated carbon fiber concentrates
having polymer sizing chosen from the aforementioned group of potential

5   polymers (i.e., polyphenylene sulfides, modified polyphenylene oxides, liquid
crystal polymers, polyamides, polyimides, polyesters, phenolics, epoxy-
containing resins, epoxy novolacs and vinyl esters).

The preferred composition contains 45-95 wt% graphite powder, 5-
50 wt% polymer resin, and 0-20 wt% metallic fiber, carbon fiber and/or

10   carbon nanofiber. Where metallic fibers are added, it is preferred that at
least 50 percent of the fibers have diameters ranging from a few nanometers
to about 50 microns, and aspect ratios ranging from 10 to 5,000.

The composition is formed into a composite having a desired
geometry by compression molding, injection molding, or a combination

15   thereof. In the case of compression molding, the graphite and polymer
powders, and/or metal-coated carbon particles or fibers, are initially blended
together to obtain a uniform distribution and composition. A preform of the
mixed blend is created by compressing the blend using a pressure of 5-
$100(10)^6$ N/m^2 at a temperature below the melting temperature of the

20   polymer constituent, and preferably at room temperature. The preform is
heated to a temperature greater than the polymer melting temperature for a
period of approximately 1-45 minutes. Subsequently, the preform is placed
between mold platens heated to a temperature in the range of 180-350°C.
The mold platens are brought together at a clamping pressure of about 1-

25    $15(10)^6$ N/m^2 and trapped gas within the mold is removed by a degassing
step in which a vacuum is applied. The degassing step takes approximately
1 minute. Following degassing, the mold clamping pressure is increased to
about $5-75(10)^6$ N/m^2. Subsequently, the mold is cooled to a temperature in
the range of approximately 80-250°C, and the part is removed from the

30   mold.

In the case of injection molding, the filler and polymer powders, and/or metal-coated carbon particles or fibers, are initially blended together to obtain a uniform distribution and composition, compounded into pellets, and then plasticized prior to injection into a mold. Where injection molding is

5   employed, the composition must be maintained at an adequate temperature (i.e., well above the melting temperature of the polymer resin) to prevent the resin from freezing, or solidifying, as it flows into and through the cooler mold. To further aid in preventing the resin from freezing during injection, the mold itself is preferably heated to a temperature of approximately 80-

10   350°C. The mixture is rapidly injected into the mold to minimize heat loss due to the high thermal conductivity of the composition. Rapid injection also produces improved material flow into and through the mold by generating higher shear forces. Varying the injection pressure can affect the rate of injection. The injection pressure may vary depending upon a number of

15   factors, such as composition viscosity, mold temperature, etc. However, it is preferred that the injection pressure is set at the maximum level that can be achieved without creating excessive mold flash. Mold flash occurs when the mold material is squeezed out of the mold cavity. Injection pressures may range from approximately $13-500(10)^6$ N/m^2. The step of injection

20   takes approximately 1-15 seconds. Following injection, the part may be retained in the mold prior to being ejected.

In a some instances, it may be desirable to employ a combination injection/compression molding process wherein the injection molded structure is subjected to a compression step following molding. This final

25   compression step may, for instance, be employed to further enhance the conductivity of the molded structure by increasing the conductive filler packing density.

In an alternate embodiment of the invention, the novel composition is melted and applied to a metallic surface to provide a hardened, highly

30   conductive protective layer upon cooling. The composition provides a means for protecting an underlying metallic structure from corrosion, while

11

precluding a significant increase in electrical resistance. Structures suitable for fuel cell applications (i.e., having properties listed in Table 1) can be formed using numerous different coating methods. For instance, a coated structure can be formed by hot-dip coating thin, stamped or etched metal

5   substrates into a melted form of the novel composition. Additional coating methods include cladding or hot roll coating a metal sheet, and subsequently hot stamping the coated surface to form a desired surface geometry.

While the preferred embodiments of the invention have been illustrated and described, it will be clear that the invention is not so limited.

10   Numerous modifications, changes, variations, substitutions and equivalents will occur to those skilled in the art without departing from the spirit and scope of the present invention as described in the claims.

We claim:

1. A composition for forming an electrically conductive polymer composite, comprising:

a non-fluorinated polymer binder having a melt viscosity of less than 1,000 Newton-seconds per square meter $(N*s/m^2)$ over a shear rate range of 1,000 to 10,000 sec^{-1}; and

a plurality of electrically conductive particles fixed in said polymer binder, said composite having a bulk conductivity of at least approximately 10 S/cm.

2. A composition as recited in claim 1, wherein said non-fluorinated polymer binder has a melt viscosity of less than 200 Newton-seconds per square meter $(N*s/m^2)$ over a shear rate range of 1,000 to 10,000 sec^{-1}.

3. A composition as recited in claim 1, wherein said non-fluorinated polymer binder comprises a thermoplastic having a thermal index of at least 45°C.

4. A composition as recited in claim 1, wherein said non-fluorinated polymer binder comprises a thermoplastic having a melting temperature of at least 90°C.

5. A composition as recited in claim 1, wherein said non-fluorinated polymer binder is chosen from the group of thermoplastic polymers consisting of polyphenylene sulfides, modified polyphenylene oxides, liquid crystal polymers, polyamides, polyimides, polyesters, phenolics, epoxy-containing resins and vinyl esters.

6. A composition as recited in claim 1, wherein said plurality of electrically conductive particles comprise carbon particles.

13

7. A composition as recited in claim 1, wherein said plurality of electrically conductive particles comprise graphite particles.

8. A composition as recited in claim 1, wherein said plurality of
5   electrically conductive particles have an average particle size in the range of about 0.1 to 200 microns, and preferably in the range of about 23 to 26 microns.

9. A composition as recited in claim 8, wherein said plurality of
10 electrically conductive particles have a BET-measured average surface area in the range of about 1 to 100 m^2/g, and preferably in the range of about 7 to 10 m^2/g.

10. A composition as recited in claim 1, wherein said plurality of
15 electrically conductive particles comprise at least 45 wt. % of said composition.

11. A composition as recited in claim 1, wherein said polymer composite comprises a current collector plate for a polymer electrolyte
20 membrane (PEM) fuel cell.

12. In a polymer electrolyte membrane (PEM) fuel cell having an ion exchange membrane and electrodes interposed between a pair of current collector plates, the improvement which comprises forming a current
25 collector plate having a composition comprising a non-fluorinated polymer having a melt viscosity of less than 1,000 N*s/m^2 over a shear rate of 1,000 to 10,000 sec^{-1}.

13. The PEM fuel cell of claim 12, wherein said current collector plate
30 comprises a unitary structure molded from said composition.

14. The PEM fuel cell of claim 12, wherein said current collector plate comprises a metal substrate coated with said composition.

15. The PEM fuel cell of claim 12, wherein said non-fluorinated polymer binder has a melt viscosity of less than 200 Newton-seconds per square meter ($N*s/m^2$) over a shear rate range of 1,000 to 10,000 sec$^{-1.}$

16. The PEM fuel cell of claim 12, wherein said non-fluorinated polymer comprises a thermoplastic resin having a thermal index of at least 45°C.

17. The PEM fuel cell of claim 12, wherein said non-fluorinated polymer comprises a thermoplastic resin having a melting temperature of at least 90°C, and preferably in the range of approximately 250 to 350°C .

18. The PEM fuel cell of claim 12, wherein said non-fluorinated polymer binder is chosen from the group of thermoplastic polymers consisting of polyphenylene sulfides, modified polyphenylene oxides, liquid crystal polymers, polyamides, polyimides, polyesters, phenolics, epoxy-containing resins and vinyl esters.

19. The PEM fuel cell of claim 12, wherein said composition further comprises a plurality of electrically conductive filler particles having an average particle size in the range of about 0.1 to 200 microns, and preferably in the range of about 23 to 26 microns.

20. The PEM fuel cell of claim 19, wherein said plurality of electrically conductive filler particles have an average surface area in the range of about 1 to 100 m^2/g, and preferably in the range of about 7 to 10 m^2/g.

15

21. The PEM fuel cell of claim 19, wherein said plurality of electrically conductive particles comprise at least 45 wt. % of said composition.

22. The PEM fuel cell of claim 12, wherein said collector plates comprise compression-molded collector plates.

23. The PEM fuel cell of claim 12, wherein said collector plates comprise injection-molded collector plates.

24. A method of fabricating a current collector plate for a polymer electrolyte (PEM) fuel cell, comprising the steps of:

combining a conductive filler powder and a nonfluorinated polymer powder to form mixture;

introducing said mixture into a cavity of a mold, said mold heated to a temperature greater than the melting temperature of said nonfluorinated polymer powder;

compressing said mixture to form a current collector plate;

cooling said mold to a temperature below the melting temperature of said nonfluorinated polymer powder; and

removing said current collector plate from said mold.

25. A method of fabricating a current collector plate for a polymer electrolyte (PEM) fuel cell, comprising the steps of:

combining a conductive filler powder and a nonfluorinated polymer powder to form mixture;

forming said mixture into a solid pellet;

heating said pellet to a temperature greater than the melting temperature of said nonfluorinated polymer powder;

injecting said mixture into a mold cavity;

allowing said mixture to cool to a temperature below the melting temperature of said nonfluorinated polymer powder to form a unitary collector plate; and
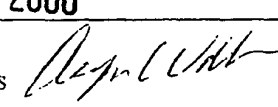
removing said unitary collector plate from said mold cavity.

5

26. The method recited in claim 25, further comprising the step of compressing said unitary collector plate.

INTERNATIONAL SEARCH REPORT

A. CLASSIFICATION OF SUBJECT MATTER
IPC(7) :H01M 8/10
US CL : 429/30
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED
Minimum documentation searched (classification system followed by classification symbols)
U.S. : 429/30

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

EAST, WEST

C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 4,197,178 A (PELLEGRI et al) 08 April 1980, col. 2, lines 35-45.	1-7 9-10
Y	US 4,851,304 A (MIWA et al) 25 July 1989, col. 3, lines 30-50.	1-7 9-10
Y,P	US 5,942,347 A (KONCAR et al) 24 August 1999, col. 6, lines 25-53.	1-26

☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

*	Special categories of cited documents:
"A"	document defining the general state of the art which is not considered to be of particular relevance
"E"	earlier document published on or after the international filing date
"L"	document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O"	document referring to an oral disclosure, use, exhibition or other means
"P"	document published prior to the international filing date but later than the priority date claimed

"T"	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X"	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y"	document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&"	document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
29 JANUARY 2000	14 FEB 2000

Name and mailing address of the ISA/US	Authorized officer
Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231	MONIQUE WILLS
Facsimile No. (703) 305-3230	Telephone No. (703) 308-0661

Form PCT-ISA/210 (second sheet)(July 1992)★